

# DUALITY AND EFFECTIVE CONDUCTIVITY OF RANDOM TWO-PHASE FLAT SYSTEMS

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The possible functional forms of the effective conductivity  $\sigma_e$  of the randomly inhomogeneous two-phase systems at arbitrary values of concentrations are discussed. Two different solutions for effective conductivity are found using a duality relation, a series expansion of  $\sigma_e$  in the inhomogeneity parameter  $z$  and some additional conjectures about the functional form of  $\sigma_e$ . They differ from the effective medium approximation, satisfy all necessary requirements, and reproduce the known formulas for  $\sigma_e$  in the weakly inhomogeneous case. This can also signify that  $\sigma_e$  of the two-phase randomly inhomogeneous systems may be a nonuniversal function, depending on some details of the structure of the random inhomogeneities.

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The electrical transport properties of the disordered systems have an important practical interest. For this reason they are intensively studied theoretically as well as experimentally. In this region there is one classical problem about the effective conductivity  $\sigma_e$  of inhomogeneous (randomly or regularly) heterophase system which is a mixture of  $N(N \geq 2)$  different phases with different conductivities  $\sigma_i, i = 1, 2, \dots, N$ . We confine ourselves here by the simplest case of the two-dimensional heterophase systems with  $N = 2$ . Despite of its relative simplicity only a few general exact results have been obtained so far. Firstly, there is a general expression for  $\sigma_e$  in case of weakly inhomogeneous isotropic medium, when the conductivity fluctuations  $\delta\sigma$  are smaller than an average conductivity  $\langle\sigma\rangle$  [1]

$$\sigma_e = \langle\sigma\rangle \left(1 - \frac{\langle(\delta\sigma)^2\rangle}{D\langle\sigma\rangle^2}\right) = \langle\sigma\rangle \left(1 - \frac{\langle\sigma^2\rangle - \langle\sigma\rangle^2}{D\langle\sigma\rangle^2}\right), \quad (1)$$

where  $D$  is a dimension of the system. In our case of two-dimensional two-phase system  $\langle\sigma\rangle = x\sigma_1 + (1-x)\sigma_2$ ,  $\langle\sigma^2\rangle - \langle\sigma\rangle^2 = 4x(1-x)\sigma_-^2$ , where  $x$  is a concentration of the first phase,  $\sigma_- = (\sigma_1 - \sigma_2)/2$ , and (1) takes a form

$$\sigma_e = \sigma_1 \left(1 - (1-x)\frac{2\sigma_-}{\sigma_1} - \frac{4x(1-x)\sigma_-^2}{2\sigma_1^2}\right) = \sigma_+ \left(1 + 2(x-1/2)z - 2x(1-x)z^2\right), \quad (2)$$

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where  $\sigma_+ = (\sigma_1 + \sigma_2)/2$ , and a new variable  $z = \sigma_-/\sigma_+$ , characterizing an inhomogeneity of the system, is introduced.

The further progress in the solution of this problem is connected with the discovery of a dual transformation, interchanging the phases [2, 3]. This transformation allows to find an exact formula for  $\sigma_e$  in the case of systems with equal concentrations of the phases  $x = x_c = 1/2$  [3]

$$\sigma_e = \sqrt{\sigma_1 \sigma_2}. \quad (3)$$

This remarkable formula is very simple and universal, since it does not depend on the type of the inhomogeneous structure of the two-phase system. For systems with unequal phase concentrations a dual transformation gives a relation between effective conductivities at adjoint concentrations  $x$  and  $1 - x$  or in terms of a new variable  $\epsilon = x - x_c$  ( $-1/2 \leq \epsilon \leq 1/2$ ) at  $\epsilon$  and  $-\epsilon$

$$\sigma_e(x, \sigma_1, \sigma_2) \sigma_e(1 - x, \sigma_1, \sigma_2) = \sigma_1 \sigma_2 = \sigma_e(\epsilon, \sigma_1, \sigma_2) \sigma_e(-\epsilon, \sigma_1, \sigma_2). \quad (4)$$

Relation (4) means that the product of the effective conductivities at adjoint concentrations is an invariant. Due to this relation, one can consider  $\sigma_e$  only in the regions  $x \geq x_c$  ( $\epsilon \geq 0$ ) or  $x \leq x_c$  ( $\epsilon \leq 0$ ).

However, an explicit formula for the effective conductivity at arbitrary phase concentrations and  $z$  attracts the main interest in this problem. One such formula was obtained many years ago in the so-called effective medium (EM) approximation [4], which turned out to be a good approximation for random resistor networks not only in the weakly inhomogeneous case [5]. In this paper, using a duality relation and a series expansion in the inhomogeneity parameter  $z$ , we will find two explicit approximate expressions for the effective conductivity of two-phase systems, differing from the EM approximation. The physical models, corresponding to them, are introduced in other papers, where their properties are discussed in detail [6, 7].

Let us start our investigation of the isotropic classical random two-phase system in the case of arbitrary concentrations with a general analysis of the possible functional form of the effective conductivity. Due to the linearity of the defining equations ([1, 3]), the effective conductivity of the random systems must be a homogeneous function of degree one of  $\sigma_i$  ( $i = 1, \dots, N$ ). In the case of  $N = 2$ , it is convenient to use, instead of  $\sigma_i$  ( $i = 1, 2$ ), the variables  $\sigma_+$  and  $z$  ( $-1 \leq z \leq 1$ ), and, instead of  $x$ , a new variable  $\epsilon$ . Then the effective conductivity can be represented in the following form, symmetrical relative to both phases,

$$\sigma_e(\epsilon, \sigma_+, \sigma_-) = \sigma_+ f(\epsilon, \sigma_-/\sigma_+) = \sigma_+ f(\epsilon, z), \quad (5)$$

where  $\sigma_e(\epsilon, \sigma_+, \sigma_-)$  and  $f(\epsilon, z)$  must have the next boundary values

$$\begin{aligned} \sigma_e(1/2, \sigma_+, \sigma_-) &= \sigma_1, & \sigma_e(-1/2, \sigma_+, \sigma_-) &= \sigma_2, \\ f(1/2, z) &= 1 + z, & f(-1/2, z) &= 1 - z, & f(\epsilon, 0) &= 1. \end{aligned} \quad (5')$$

The duality relation in these variables takes the form

$$f(\epsilon, z) f(-\epsilon, z) = 1 - z^2, \quad (6)$$

from which it follows that at critical concentration  $\epsilon = 0$

$$f(0, z) = \sqrt{1 - z^2}. \quad (3')$$

Strictly speaking, the form of a duality relation (6) is also a consequence of another exact relation for the effective conductivity, taking place at arbitrary concentrations for systems with the similar random structures of both phases of the system,

$$\sigma_e(\epsilon, \sigma_1, \sigma_2) = \sigma_e(-\epsilon, \sigma_2, \sigma_1). \quad (7)$$

This means that the effective conductivity of the random two-phase system must be invariant under substitution of these phases ( $\sigma_1 \longleftrightarrow \sigma_2$ ) with the corresponding change of their concentrations  $x \longleftrightarrow 1 - x$  (or  $\epsilon \rightarrow -\epsilon$ ). In the new variables this means that

$$f(\epsilon, z) = f(-\epsilon, -z), \quad f(-\epsilon, z) = f(\epsilon, -z). \quad (8)$$

For this reason, the duality relation can also be written in the form

$$f(\epsilon, z)f(\epsilon, -z) = 1 - z^2. \quad (9)$$

It follows from (8) that the even ( $f_s$ ) and odd ( $f_a$ ) parts of  $f(\epsilon, z)$  relative to  $\epsilon$  coincide with the even ( $f^s$ ) and odd ( $f^a$ ) parts of  $f(\epsilon, z)$  relative to  $z$ . Consequently,  $f(\epsilon, z)$  has the functional form

$$f(\epsilon, z) = f(\epsilon z, \epsilon^2, z^2). \quad (10)$$

It follows from (10) that: (1)  $f(0, z)$  is an even function of  $z$  (i.e. symmetric in  $\sigma_{1,2}$ ), (2) an expansion of  $f(\epsilon, z)$  near the point  $\epsilon = z = 0$  does not contain terms linear in  $\epsilon$  and  $z$  separately. Analogously, the odd part  $f_a$  can be represented in the form

$$f_a(\epsilon, z) = 2\epsilon z \Phi(\epsilon, z), \quad (11)$$

where  $\Phi$  is an even function of  $\epsilon$  and  $z$  (the coefficient 2 in front of  $\epsilon z$  is chosen for further convenience).

At first sight, the duality relation (11) alone is not enough for the complete determination of  $f$  in the general case. It only connects  $f$  at adjoint concentrations or  $f_a$  and  $f_s$

$$f_s^2 - f_a^2 = 1 - z^2. \quad (12)$$

This means that  $f_a$  and  $f_s$  considered at fixed  $z$  as the functions of  $\epsilon$  satisfy the hyperbolic relation with a constant depending on  $z$ . The relation (12) allows one to express  $f(\epsilon, z)$  through its even or odd parts

$$f(\epsilon, z) = f_a + \sqrt{f_a^2 + 1 - z^2} = f_s \pm \sqrt{f_s^2 - 1 + z^2}. \quad (13)$$

For this reason, it is enough to know only one of these two parts. Usually, one prefers to choose an antisymmetric part as a more simple one. It follows from

(2) that, in the weakly inhomogeneous case, the odd part coincides with the odd part of  $\langle\sigma\rangle$  and has the simplest form (compatible with (11))

$$f_a(\epsilon, z) = 2\epsilon z. \quad (14)$$

As is well known, the effective conductivity in the EM approximation can be obtained by substitution of (14) into (13):

$$\sigma_e(\epsilon, z) = \sigma_+ \left[ 2\epsilon z + \sqrt{(2\epsilon z)^2 + 1 - z^2} \right]. \quad (15)$$

We will call this expression, continued on arbitrary concentrations  $x = \epsilon + 1/2$  and inhomogeneties  $z$ , the EM approximation for  $\sigma_e$ .

However, systems with a dual symmetry usually have some additional hidden properties, permitting one to obtain more information about function under question. Moreover, in some cases these properties can help to solve problem exactly (see, for example, [8]). Having this in mind, we will try to investigate the duality relation in more detail. For every fixed  $z \neq 1$  (it is enough to consider only the region  $0 \leq z \leq 1$ ), a function  $f$  must be a monotonous function of  $\epsilon$ . Since the homogeneous limit  $z \rightarrow 0$  is a regular point of  $f$ , it will be very useful to expand  $f$  in a series in  $z$

$$f(\epsilon, z) = \sum_0^{\infty} f_k(\epsilon) z^k / k!, \quad (16)$$

where due to the boundary conditions (5')

$$f_0 = 1, \quad f_1(\epsilon) = 2\epsilon. \quad (17)$$

It is worth noting here that the expansion (16) differs from the weak-disorder expansion of  $\sigma_e$  in series on the averaged powers of the conductivity fluctuations  $\delta\sigma/\langle\sigma\rangle$  (see, for example [9]). Expansion (16) is simpler, since it deals with variables  $z$  and  $\epsilon$  separately, while the expansion in powers of  $\delta\sigma/\langle\sigma\rangle$  is an expansion on the rather complicated functions of  $z$  and  $\epsilon$ . Of course, both expansions are connected, but expansion (16) is more convenient for our analysis of possible functional forms.

Substituting the expansion (16) into (6) one obtains the following results:

(1) in the second order on  $z$  it reproduces a universal formula (2), thus the latter can be considered as a consequence of the duality relation;

(2) in higher orders there are recurrent relations between  $f_{2k}$  and  $f_{2k-1}$ , corresponding to connection (12);

(3)  $f_{2k+1}(\epsilon)$  are odd polynomials in  $\epsilon$  of degree  $2k+1$  and  $f_{2k}(\epsilon)$  are even polynomials in  $\epsilon$  of degree  $2k$  in agreement with (10).

Taking into account boundary conditions (5') and an exact value (3'), one can show that the coefficients  $f_k$  must have the next form

$$\begin{aligned} f_{2k+1}(\epsilon) &= \epsilon(1 - 4\epsilon^2)g_{2k-2}(\epsilon), \quad k \geq 1, \\ f_{2k}(\epsilon) &= (1 - 4\epsilon^2)h_{2k-2}(\epsilon), \quad k \geq 1, \end{aligned} \quad (18)$$

where  $g_{2k-2}$  and  $h_{2k-2}$  are some even polynomials of the corresponding degree and free terms of  $h_{2k-2}$  coincide with the coefficients in the expansion of (3')

$$\sqrt{1-z^2} = 1 - z^2/2 - z^4/8 - z^6/16 - z^8/128 - z^{10}/256 + \dots \quad (19)$$

It follows from (18) that  $f_3$  is completely determined up to overall factor number  $g_0$ . Since  $f_4$  is determined through lower  $f_k$  ( $k = 1, 2, 3$ )

$$f_4 = 4f_1f_3 - 3f_2^2 = (1 - 4\epsilon^2)[(8g_0 + 12)\epsilon^2 - 3], \quad (20)$$

it is also determined by the coefficient  $g_0$ . Expansion (16) in the EM approximation has a very simple form, since all  $g_{2k-2} = 0$  ( $k \geq 1$ ) and  $f_{2k}(\epsilon) \sim (1 - 4\epsilon^2)^k$ . Thus, we see that, in general case, the arbitrariness of  $f$  is strongly reduced by boundary conditions and by exact value (3) and that the third and fourth orders are determined only up to one constant. One can see from the EM approximation that any additional information about function  $f$  can determine this constant or even the whole function. For this reason one needs to know what kind of functions can satisfy the duality relation (6) except general functions from (12),(13). In order to answer this question it is convenient in the case  $z \neq 1$  to pass from  $f$  to  $\tilde{f} = f/\sqrt{1-z^2}$ . Then

$$\tilde{f}(\epsilon, z)\tilde{f}(-\epsilon, z) = 1 = \tilde{f}(\epsilon, z)\tilde{f}(\epsilon, -z). \quad (6')$$

The duality relation gives some constraints on the possible functional form of  $\tilde{f}(\epsilon, z)$ . For example, assuming a functional form (10), one can write out the next simple expression:

$$\tilde{f}(\epsilon, z) = \exp(\epsilon z \phi(\epsilon, z)), \quad (21)$$

where  $\phi(\epsilon, z)$  is some even function of its arguments. Another possible form of  $\tilde{f}$  is

$$\tilde{f}(\epsilon, z) = B(\epsilon z)/B(-\epsilon, z).$$

It is easy to see that they automatically satisfy eq.(6').

Let us now consider two simple ansatzes for a function  $\phi$ . In case (a) we suppose that  $\phi(\epsilon, z)$  depends only on  $z$ . This means an exponential dependence on concentration, which sometimes takes place in disordered systems [10]. In case (b) we will suppose that  $\phi(\epsilon, z)$  depends only on the combination  $\epsilon z$ . This can signify, for example, that  $f$  depend only on mean conductivity  $\langle\sigma\rangle$  and/or on mean resistivity  $\langle\sigma^{-1}\rangle$ , since  $\langle\sigma^\pm\rangle \sim (1 \pm 2\epsilon z)$ . Expanding the corresponding functions  $\tilde{f}$  in series, one can check after some algebra that it is now possible to determine all polynomial coefficients unambiguously! For example, one finds for  $f_a$  in the 3-d and 5-th orders

$$\begin{aligned} g_0 &= -1, & g_2 &= -(11 + 4\epsilon^2), & \text{case(a)}, \\ g_0 &= -3, & g_2 &= -15(1 + 12\epsilon^2), & \text{case(b)}. \end{aligned}$$

Another way to see this is to apply boundary conditions directly to the function (21). In the case (a) one obtains

$$\phi(z) = 1/z \ln \frac{1+z}{1-z}, \quad \tilde{f}(\epsilon, z) = \left( \frac{1+z}{1-z} \right)^\epsilon. \quad (22)$$

It is interesting to note that, in terms of concentration  $x$  and partial conductivities  $\sigma_i$ , one obtains in this case

$$\sigma_e = \sigma_1^x \sigma_2^{1-x}. \quad (22')$$

This corresponds to the self-averaging of  $\ln \sigma$  :

$$\sigma_e = \exp\langle\sigma\rangle, \quad \langle\sigma\rangle = x \ln \sigma_1 + (1-x) \ln \sigma_2,$$

noted first by Dykhne for the case of equal phase concentrations [3] and established later in the theory of weak localization [11].

In case (b), when  $\phi$  depends only on the combination  $\epsilon z$ , one finds

$$\phi(\epsilon z) = \frac{1}{2\epsilon z} \ln \frac{1+2\epsilon z}{1-2\epsilon z}, \quad \tilde{f}(\epsilon, z) = \left( \frac{1+2\epsilon z}{1-2\epsilon z} \right)^{1/2}. \quad (23)$$

In terms of  $x$  and  $\sigma_i$  it has the next simple form

$$\sigma_e = \sqrt{\langle\sigma\rangle/\langle\sigma^{-1}\rangle}. \quad (23')$$

Series expansions of (22) and (23) coincide exactly with the corresponding expansions mentioned above. They differ from the EM approximation already in the third order.

For a general form of  $\phi(\epsilon, z)$ , admitting a double series expansion in  $z^2$  and  $\epsilon^2$ ,

$$\phi(\epsilon, z) = \sum_0^\infty \phi_k(\epsilon) z^{2k}/k!, \quad \phi_k(\epsilon) = \sum_0^\infty \phi_{kl} \epsilon^{2l}/l!,$$

one can show that now  $f_3$  and  $f_4$  again contain one free parameter  $\phi_{10}$  :  $g_0 = 6(\phi_{10} - 1)$ . Consequently, one needs additional information or a more complicated ansatz for a determination of  $\phi$  in the general case. This will be considered in another paper.

Thus we have found two explicit functions (22) and (23), which satisfy all required properties. In particular, they reproduce equation (2) in the weakly inhomogeneous limit  $z \ll 1$ . These functions can be considered as regular solutions of the duality relation, since they are represented by convergent series in  $z$  for  $0 \leq z \leq 1$  except the small region  $z \rightarrow 1, \epsilon \rightarrow 1/2$ .

The systems, having the effective conductivity just of two forms found above and their properties are considered in the other paper [6] (see also [7]). We give here only their brief description.

The first model represents randomly inhomogeneous systems with compact inclusions of the second phase with finite maximal scale  $l_m$  of inhomogeneities.

This scale can depend on concentration of the second phase  $l_m(1-x)$  (one can consider only the case  $1-x \leq 1/2$ ). The stable effective conductivity  $\sigma_e(x, \{\sigma\})$  (here  $\{\sigma\} = (\sigma_1, \sigma_2)$ ), depending only  $x$  and not depending on the scale on which the averaging is performed, can be obtained only after averaging over scales  $l > l_m(x)$ . This  $\sigma_e(x, \{\sigma\})$  as a function of  $x$  must satisfy the next functional equation, generalizing duality relation (4)

$$\sigma_e(x', \{\sigma\})\sigma_e(x'', \{\sigma\}) = \sigma_e^2(x, \{\sigma\}), \quad (24)$$

where  $x = (x' + x'')/2$ . The solution of equation (24), satisfying boundary conditions (5'), coincides with (22) and corresponds to the finite maximal scale averaging approximation (FMSA) [6, 7].

The second model of a random inhomogeneous systems has a hierarchical, two-level structure. On the first level, it consists of squares with random phase layers with a mean conductivity  $\langle\sigma\rangle$  if the direction of layers is parallel to the applied electrical field  $\mathbf{E}$  or with a conductivity  $\langle\sigma^{-1}\rangle^{-1}$  if this direction is perpendicular to  $\mathbf{E}$ . On the second level, these squares form a random parquet (or a lattice), which contains with equal probabilities ( $p = 1/2$ ) squares with both orientations. Then, using universal formula (3), one can write the next approximate expression for  $\sigma_e$

$$\sigma_e(x, \{\sigma\}) = \sqrt{\langle\sigma\rangle\langle\sigma^{-1}\rangle^{-1}},$$

which coincides with (23).

For a comparison of the different expressions for effective conductivity (eqs. (15), (22) and (23)), we have constructed three plots of the corresponding functions  $f(\epsilon, z)$  at  $z = 0.8, 0.95, 0.999$  (Fig.1) (their full 3D plots are represented in [7]). The lower branch in the region  $\epsilon > 0$  corresponds to  $f$  from (23), the upper branch to the EM approximation, and the middle branch to  $f$  from (22). It appears that all three formulas for  $f(\epsilon, z)$ , despite of their various functional forms, differ from each other very weakly for  $z \lesssim 0.5$  due to very restrictive boundary conditions (5') and the exact Keller-Dykhne value. This range of  $z$  corresponds approximately to the ratio  $\sigma_2/\sigma_1 \sim 1/3$ . For the smaller ratios, the differences between these functions become distinguishable (for  $\epsilon > 0$ ), growing significantly only for ratios  $\sigma_2/\sigma_1 \lesssim 10^{-1}$ .

One can see from formulas (22), (23) that, in both cases, one gets  $\sigma_e \rightarrow 0$  in the limit  $\sigma_2 \rightarrow 0$ , except the small region near  $x = 1$  and  $z = 1$ . This means that these formulas are not valid in the percolation limit  $\sigma_2 \rightarrow 0$  ( $z \rightarrow 1$ ) for  $\epsilon > 0$  [10, 12]. One can show that such behaviour is a consequence of the assumptions made about the form of the function  $\phi$  and/or of the structure of the corresponding models [7].

This can be connected also with a possible divergence of the series (16) in general case, when  $z \rightarrow 1$ , due to a singular behaviour of  $\sigma_e$  in the percolation problem [9, 10].

For this reason the formulas (22, 23) cannot be applicable for the description of  $\sigma_{eff}$  in the limit  $z \rightarrow 1$  ( $\epsilon > 0$ ) and the corresponding percolation problem. It follows also from the constructed plots that EM approximation overestimates

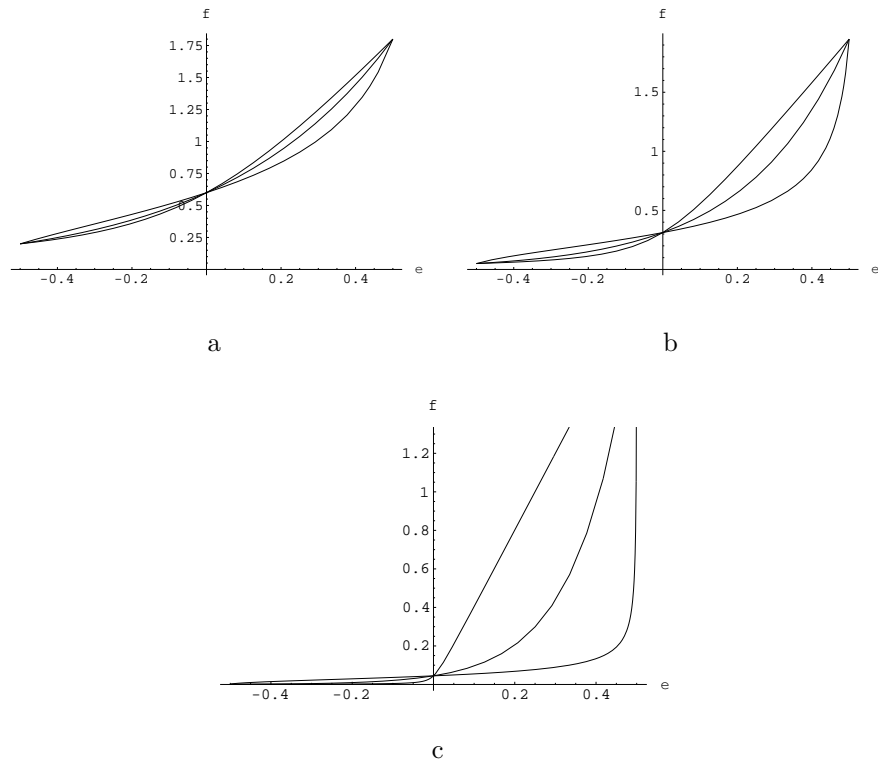


Fig.1. Plots of various expressions for  $f(\epsilon, z)$  at: (a)  $z = 0.8$ , (b)  $z = 0.95$ , (c)  $z = 0.999$ .

$\sigma_e$  [10, 12], and both the other formulas underestimate it in the region  $z \rightarrow 1, \epsilon > 0$ . We hope to investigate this limit in detail later.

Thus, we have discussed possible functional forms of the effective conductivity of random two-phase systems at arbitrary values of concentrations. It was shown that the duality relation and some additional assumptions about possible functional form of  $f(\epsilon, z)$  can give its explicit expressions, differing from EM approximation. They automatically satisfy the duality relation and reproduce all known formulas for  $f$  in the weakly inhomogeneous limit  $z \ll 1$ .

Though the used additional assumptions are the approximate ones the obtained results (and especially an existence of the corresponding models [6, 7]) can be interpreted also as if  $\sigma_e$  of the two-phase randomly inhomogeneous systems were a nonuniversal function, depending on some details of the structure of the random inhomogeneities. An analogous conclusion was made earlier for three-phase *regular* systems in [13], where a possibility to find a generalization of the Keller – Dykhne formula (3) for the case  $N = 3$  was studied numerically.

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